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June 28, 2011

INMM 52nd Annual Meeting  
Palm Desert, CA, United States  
July 17, 2011 through July 21, 2011

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# **Improving Turn Around Times Of Calorimeter Measurements In The LLNL MC&A Labs Through Reduction Of Thermal Impedance**

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## **Abstract**

The LLNL MC&A program has experienced a nearly two-fold increase in measurement demand since September of 2007. This influx is due to the NNSA decision that LLNL de-inventory its Safeguards Category I/II nuclear material no later than September 30, 2012. Adding new measurement equipment was considered, but the requisite procurement, installation, and certification processes would not have been completed in time to have a sufficient impact. Further, such an effort would have diverted the time and attention of specialized personnel away from the de-inventory effort itself. Instead, measurement throughput was increased largely through overtime and meticulous workflow planning. Strategic adaptations to existing techniques and equipment provided the remaining gains in throughput that enabled all of the de-inventory milestones to be met or exceeded.

For calorimeter measurements we sought ways to reduce run-times by speeding the time to thermal equilibrium. We accomplished this by minimizing the thermal resistance of our packing material and striving to improve conduction. The result was an overall increase of 25% in the number of calorimeter runs. This was significant since most items measured during the de-inventory period required calorimetry. We continue to strive for maximum speed with investigations into configurations that facilitate the end-point prediction method through improved heat transfer. We describe our experiences to optimize heat-flow and dub it our quest for, "Positive Internal Heat Goodness."

## **Background**

Safeguards of Special Nuclear Materials (SNM) is the primary function of the LLNL Materials Management Section. As such, it is responsible for tracking all accountable quantities of SNM in its inventory and demonstrating that said material is present using the principles of Materials Control and Accountability (MC&A). A crucial component is the ability to perform non-destructive assay (NDA) of SNM. Materials Management has several laboratories dedicated to performing these measurements. The measurement apparatus is maintained and calibrated by professional staff and the precision and accuracy of each instrument is tracked and validated against measurement control charts on a routine basis. The DOE Livermore Site Office

works closely with LLNL to assure that all measurement requirements are met as a condition of Livermore's license to work with SNM.

As with most measurements laboratories throughout the DOE complex, Livermore relies heavily on the use of calorimeters to perform NDA of fissile material by measuring the heat output of containerized radioactive samples. When combined with the specific heats of the various radioisotopes present, the mass for each can be derived with great accuracy. Because Livermore is a research environment there are a wide range of items in its inventory and a variety of calorimeter configurations are therefore needed to address all MC&A requirements. There are three large volume calorimeters, and four more with small volumes. Some are twin-bridge water bath designs, and others are gradient bridge air bath. Only two address the same volume and heat ranges. For many years LLNL has readily met safeguards requirements in inventory tracking. However, the demands of the de-inventory project have compelled us to increase measurement throughput.

### **De-Inventory Drives Need for Increased Throughput**

In the fall of 2007 the process of down-grading the Cat I/II nuclear facilities at LLNL to Cat III commenced as mandated by the NNSA. The deadline for achieving this was September, 2012 but with pressure from the NNSA Director's office to complete the process sooner if possible. Lawrence Livermore National Security, the organization that manages LLNL, was given the enhanced performance objective that all intermediate de-inventory milestones be met. To carry out de-inventory, LLNL needed to reconfigure nearly all of its SNM inventory into shipping friendly forms, and then repack it into containers specifically approved for transport. A massive effort therefore ensued to prepare the existing materials, per these requirements. As a consequence of this surge, the needs for safeguards measurements skyrocketed, and finding ways to accelerate measurement throughput became imperative. There was neither enough time nor personnel to select, install, qualify, and calibrate new measurement equipment in time to meet the aggressive de-inventory milestones. Instead we focused on finding ways to increase the number of measurements we could perform using existing resources. We scripted A measurement plan each day to maximize efficiency and eventually extended our work week to include off-hours on weekend mornings. However, careful planning and working weekends were not enough when a large influx of measurements came to us in a short time and created a backlog that threatened our ability to stay on schedule. Missing any of the de-inventory milestones was not an option, so we focuses our efforts on finding a way to speed up the most time critical measurement path in our labs – calorimetry.

### **Measuring Heat**

Only two of Livermore's seven calorimeters have the same design and sample capacity. Despite this they all have the same fundamental features that allow them to measure heat, so techniques that improve the performance of one can be extrapolated to the others as well.

Calorimeters measure the amount of thermal energy from a sample or 'heat', symbol  $Q$ . This is accomplished by determining the kinetic energy (KE) imparted to a substance of known thermal resistance,  $R_{Th}$ . The amount of KE is proportional to the difference in temperature readings ( $\Delta T$ ) on either side of the resistance when the flow of energy has reached a dynamic equilibrium (EQ). To quantify this, the sample is placed in a chamber that isolates it between the resistance and an outer environment that is temperature controlled. The outer environment serves as an infinite sink for heat flowing from the inner chamber through the thermal resistance. This energy biased system assures that sample heat will flow from the inside out, (Figure 1).

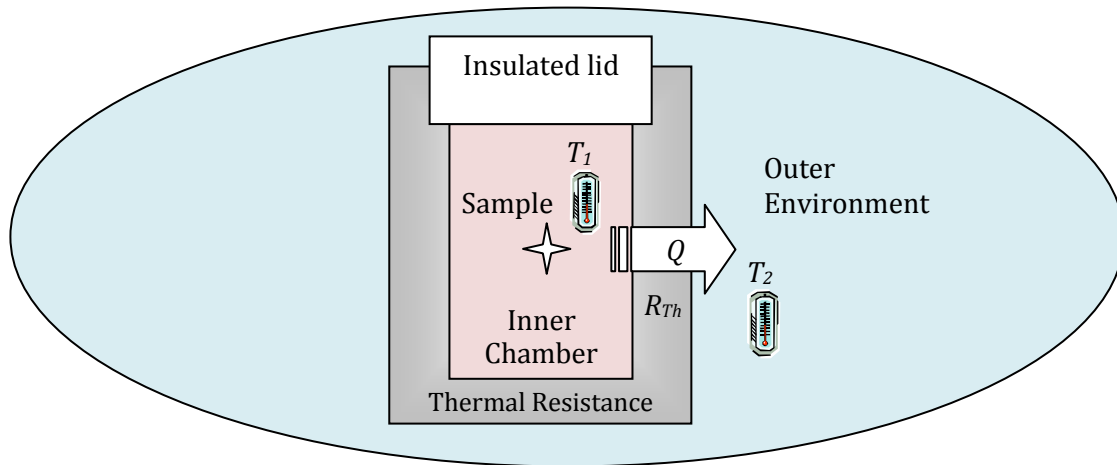


Figure 1. Schematic of a calorimeter: an inner chamber for the sample, a well defined thermal resistance, a temperature controlled outer environment, sensors indicating the temperatures on either side of the resistance, and an insulated lid

When the sample is first introduced, the KE of the inner chamber is disturbed and  $\Delta T$  changes until a new dynamic equilibrium is reached. When the system achieves a uniform temperature gradient across the thermal resistance element, then  $\Delta T$  is constant and represents the amount of heat given off by the sample, (Figure 2).

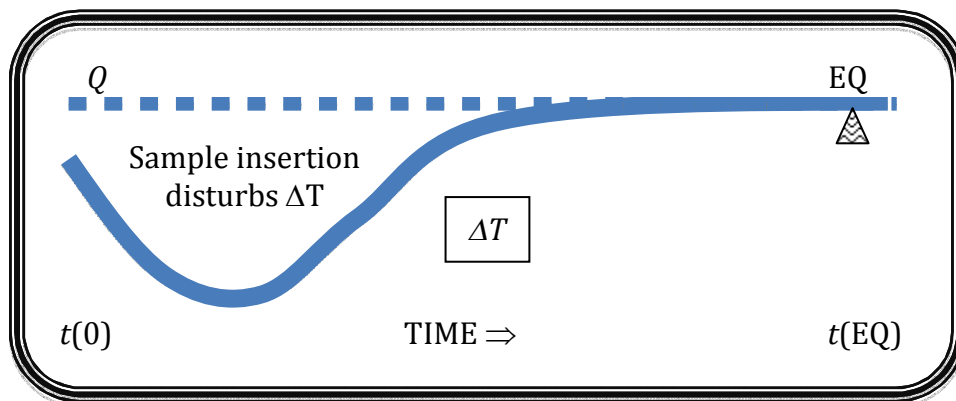


Figure 2. Sample pushes heat constantly but  $\Delta T$  changes until equilibrium

The change in heat required to achieve a particular change in temperature is dependent upon the amount of matter contained in the thermal resistance. This is specific heat capacity, symbol  $c$ . Its units are thermal energy in calories, btus, or joules, per unit mass, per °C or °K.

$$c = \frac{1}{m} \frac{dQ}{dT}$$

Rearranging ...

$$dQ = mc dT$$

Integrating, we obtain the expression for total heat required ...

$$Q = m \int_{T_1}^{T_2} c dT$$

The specific heat capacities of all materials vary somewhat as a function of temperature so  $c$  must be expressed as a function of  $T$  to carry out the integration. For solids,  $c$  doesn't change between 0 and 100 °C, and since our calorimeters operate well within this range it can be treated as a constant ...

$$Q = mc \int_{T_1}^{T_2} dT$$

The integral for heat across the resistance then becomes...

$$Q = mc (T_2 - T_1)$$

The product of mass and specific heat capacity ( $mc$ ) characterizes the body of thermal resistance itself. Its units are expressed in energy per degree (i.e., kCal/°C). We can modify our equation for heat-flow to resemble the formula for the flow of electrical current, ( $I = E/R$ ) by substituting  $1/R_{Th}$  for  $mc$  and  $\Delta T$  for  $(T_2 - T_1)$ .

$$Q = \frac{\Delta T}{R_{Th}}$$

For our calorimeters  $\Delta T$  is sampled over a finite time interval so we measure energy per unit time. The units are thermal Watts (kCal / °C s), and the symbol is  $H$ :

$$H = \frac{\Delta T}{\Delta t R_{Th}}$$

## Thermal Impedance - Resistance, Geometry and Contact

The time it takes for a calorimeter to reach thermal equilibrium depends on its ability to transfer heat. The bulk thermal resistance of matter plays a major role but other factors must also be addressed. The geometry of materials effects the average distance through which the heat propagates so that irregular shapes extend the amount of time it takes for the thermal gradient to stabilize after a disturbance. Poor contact between the various components reduces the efficiency of transfer between the various elements as well. All three factors can delay heat transfer and we refer to their combined effect informally as thermal impedance, (Figure 3).

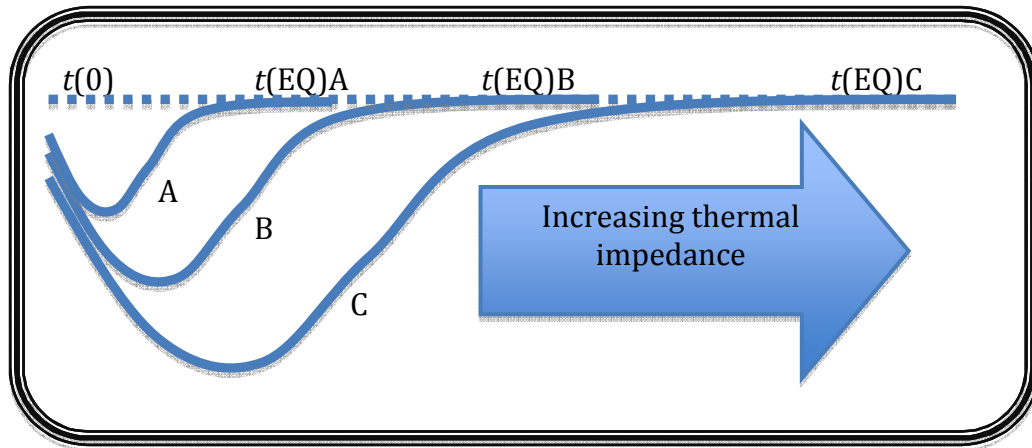


Figure 3. Increasing thermal impedance delays time to achieve equilibrium

The lag time caused by thermal impedance can be reduced by a technique called preheating, where the sample is conditioned in advance of measurement. Here, the item is soaked in a surrogate chamber to establish equilibrium before it is placed into a calorimeter. Unfortunately, the time it takes to adequately preheat is similar to the time it takes to make a 'cold' calorimeter run so unless there is sufficient lead-time to perform this operation, its value as a time saver is diminished. During de-inventory there was often no time for preheating due to the short turnaround times mandated for specific items. Also, the effectiveness of preheating is lessened if the sample chamber to be used is still warm from a previous run. In fact, reducing thermal impedance in samples is advantageous for shortening preheat times as well.

Most parts of a calorimeter are fixed and permanent but the sample, its container with internal packaging, and the packing material used to position it in the loading canister vary for each measurement. Improving the sample-packaging configuration was therefore identified as a possible way to gain time. Whatever we could do to reduce thermal impedance would benefit all our calorimeter operations including preheating. It would be Positive Internal Heat Goodness, (PIhGs). However, for PIhGs to be of value it needed to provide gains sufficient to increase measurement turnaround times in calorimeter measurements to more than our previous average of once a day. At least one of the two runs needed to be made in eight hours or less.

## Reducing Thermal Resistance

We have already discussed how the masses and specific heat capacities of matter impacts thermal resistance. However, the calorimeter model previously described does not address the resistance of the sample and its associated hardware. A cylindrical metal cavity forms the inner wall of the sample chamber. It is a fixed member of the calorimeter body and shares contact with the inner temperature sensors so it is incorporated into the calibration for  $R_{Th}$ . Nested inside of the cavity is a removable canister for loading and unloading the sample. Inside of the loading canister there is packing material that surrounds the sample container. Innermost is the radioactive sample material itself swathed in more packaging inside the sealed metal container. An insulated lid allows for access, (Figure 4).

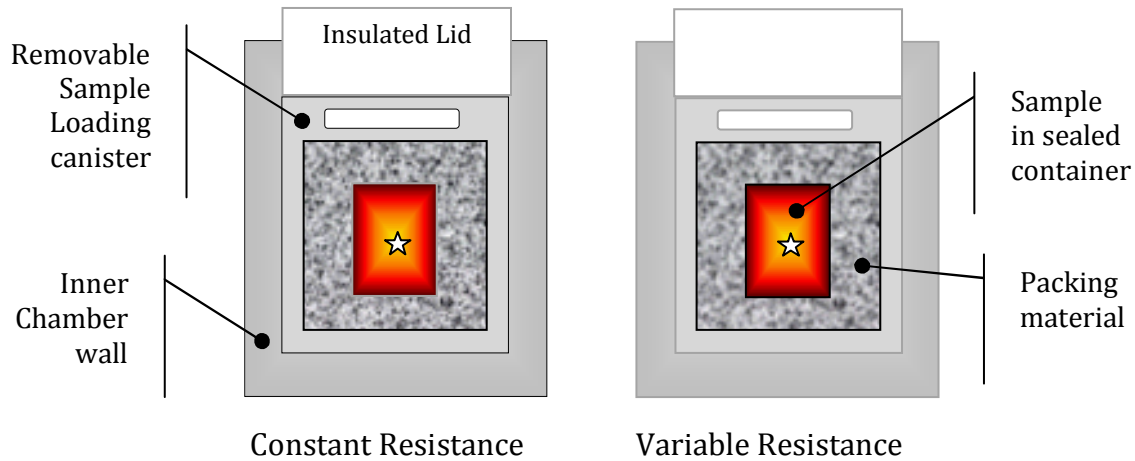


Figure 4. Anatomy of the calorimeter sample chamber – constant and variable resistances

The calculation of heat-flow requires a known, unchanging value of  $R_{Th}$  and does not include the resistances of the sample hardware. However, the concept of thermal resistance is useful in understanding how the sample hardware might be changed to speed the flow of heat toward  $R_{Th}$ . Since the direction of flow is the same, it is reasonable to treat them as additive, and that they are also inversely related to  $H$ :

$$\frac{1}{R_{Total}} = \frac{1}{R_{Th}} + \frac{1}{R_{canister}} + \frac{1}{R_{packing}} + \frac{1}{R_{sample}}$$

The thermal resistance of the sample loading canister remains the same for each measurement. However, the samples vary each time. The amount of canister packing material required therefore varies depending on the size of the sample container. Neither the sample, nor its canister packing is built into the value for  $R_{Th}$ . We have little say in how the sample is packaged but, the resistance for packing material is fair game for optimization.



## **Testing the Impact of Packing Material Part One – Zero-Powers**

When de-inventory began, the standard procedure for sample loading at LLNL was to surround the sample container in the canister with packing material. We needed to get an idea of the resistance the sample packing material was contributing, so a test was performed. This was done in two steps. First, a measurement was made with heat standards equivalent to the highest allowable for the calorimeter used. The canister was backfilled with packing material as well. When the run was complete, the standards were removed but the packing material was left in the canister and quickly placed back into the calorimeter. A follow up run was then made with the warm packing material. The time it took to pull out the heat stored in the packing material and complete this zero-power (ZP) measurement was 12 hours which was comparable to other zero-power times for that calorimeter. The calorimeter chosen for this test was one of two identical units and the twin typically required about 12 hours to complete a zero-power run as well.

The standard run was repeated and a zero-power measurement was made immediately afterward as before. This time the canister packing material was removed so that an empty canister was measured. The empty canister zero-power run completed in approximately 7 hours – a savings of 5 hours. This indicated that the resistance due to the packing material was a significant factor in measurement time. More importantly, the time to make a zero-power run was shown to be shorter than the standard work day. The significance of this was that we could make zero-power measurements in short enough time to start and finish a run in one shift. Since we run zero-powers every other day on these two systems the gain in throughput for them was immediately increased from one run every day to three runs every two days.

Similar tests were made on our other calorimeters with time gains realized for them as well. For three more, the zero-power run times were shortened from 24 hours to between 12 and 16. Although this did not allow for same-shift turnaround, it did make it possible to start a run late in the afternoon and have it finish in time to start a new sample or standard run the following morning. The accuracy of the test results for running zero-power measurements with empty canisters were validated by comparison to statistical control charts for all seven of our calorimeters.

## **Testing the Impact of Packing Material Part Two – Heat Standards**

For zero-power runs, improving thermal impedance was straight forward – just run empty. This also indicated that the reduction of packing material was a prime factor in improving run times. For standards and samples, complete elimination of packing material was not a feasible approach so the reduction of packing resistance needed to be addressed.

For the larger volume machines we were packing samples in the canisters with balls made of crumpled aluminum foil. In the small volume calorimeters we were using aluminum shot. We researched a wide variety of substitutes for both the balls and shot. These included some metallic foams, and honey comb materials that could reduce the effective density of the aluminum and hence the mass. However, to use these effectively, each would need to have been custom-made to fit the wide variety of sample containers in our inventory. It would be difficult to manage the dozens of shapes and sizes that would have been needed. Assuring that good contact was achieved would have required close fits and we worried that these materials were likely to damage the tamper indicating devices and other labeling on the sealed metal sample and standards containers.

We considered replacing the aluminum shot with copper shot but quickly saw that the thermal resistance for copper shot in the same volume would be double. Recall that the thermal resistance of matter is the product of mass and specific heat capacity. Although the specific heat of copper is only 60% of aluminum, it is also 330% more dense so that for the same volume of material aluminum transfers heat twice as fast. Comparisons with other materials showed that aluminum was already an excellent material for packing in terms of performance, availability, and cost.

Once again we needed an idea of how much we could gain by reducing the mass of the packing material we were already using. This time we tested the highest and lowest heat combinations of Pu-238 heat standards. The tests were performed with and without packing material in the canister. Each standard was preceded by a zero-power run to assure that there was no heat in the canister at time zero,  $t(0)$ , and the full effect of the packing would be evident. We used Cal 33, one of our low volume calorimeters.

For Cal 33, the sample canister can hold 3 Kg of aluminum shot. When we removed the shot, the time to reach EQ was reduced from 20 hours to 7 for our lowest wattage standard - 0.08 Watts. When we made a run at 5.5 Watts, the highest wattage standard set for this Cal, the time dropped from 23 down to 15 hours. We also made a zero-power run with an empty canister and compared it with another one of a canister filled with shot. For the empty canister the zero-power run reached equilibrium 13 hours sooner with an EQ of 7 hours versus 20, (Figures 4, 5 and 6). Each run was preceded by a zero-power, and the packing material used was at room temperature before loading. With or without the aluminum shot all results were within the statistical limits of error in our control charts.

Clearly, efforts to improve measurement times were readily possible by simply limiting the amount of material used for packing. In fact, it looked like achieving our goal of twice-daily calorimeter runs was already within our grasp. Even for the high heat runs the time to EQ was reduced from 20 to 24 hours to 12 to 16 hours, on average. As previously mentioned, completion times of less than 2/3<sup>rd</sup> day can result in gains to measurement throughput because it allows that a run started before leaving in the evening can be finished upon arrival the next morning.

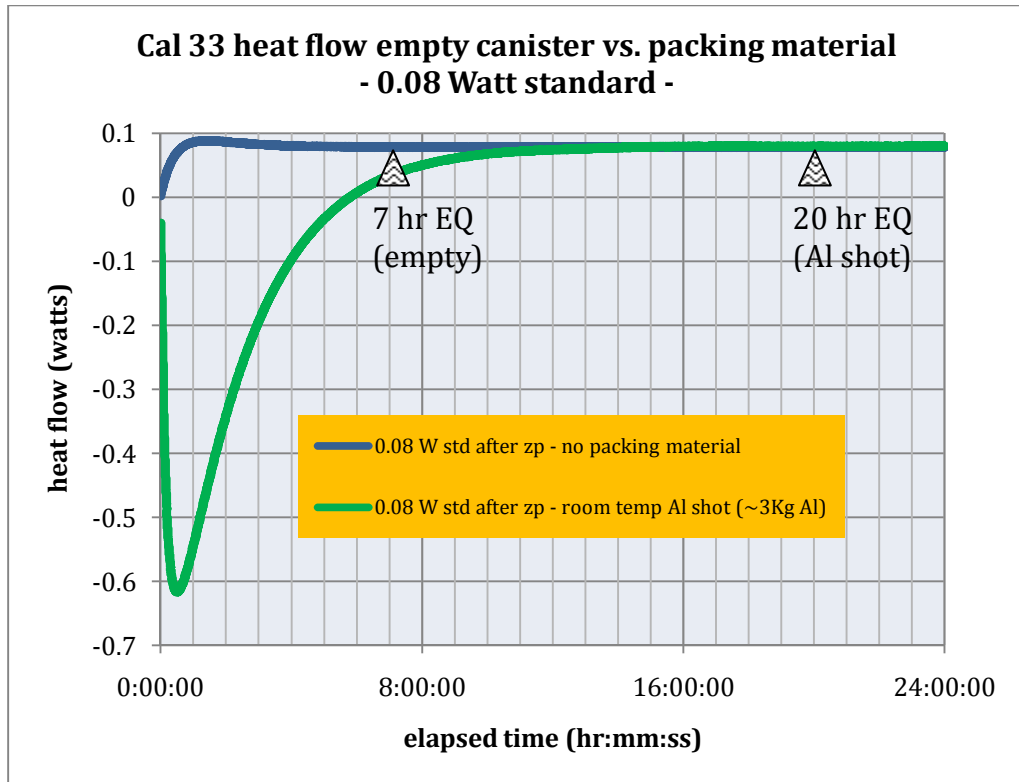


Figure 5. EQ time reduced 13 hours with 0.08 Watts for empty canister vs. packing

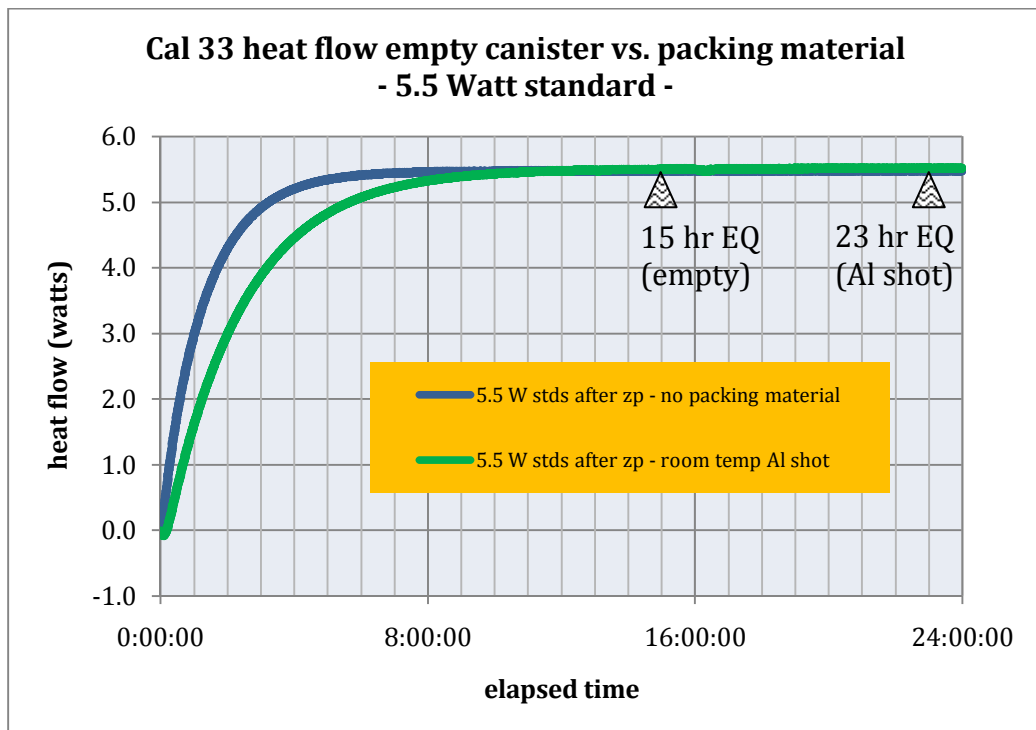


Figure 6. EQ time reduced 8 hours with 5.5 Watts for empty canister vs. packing

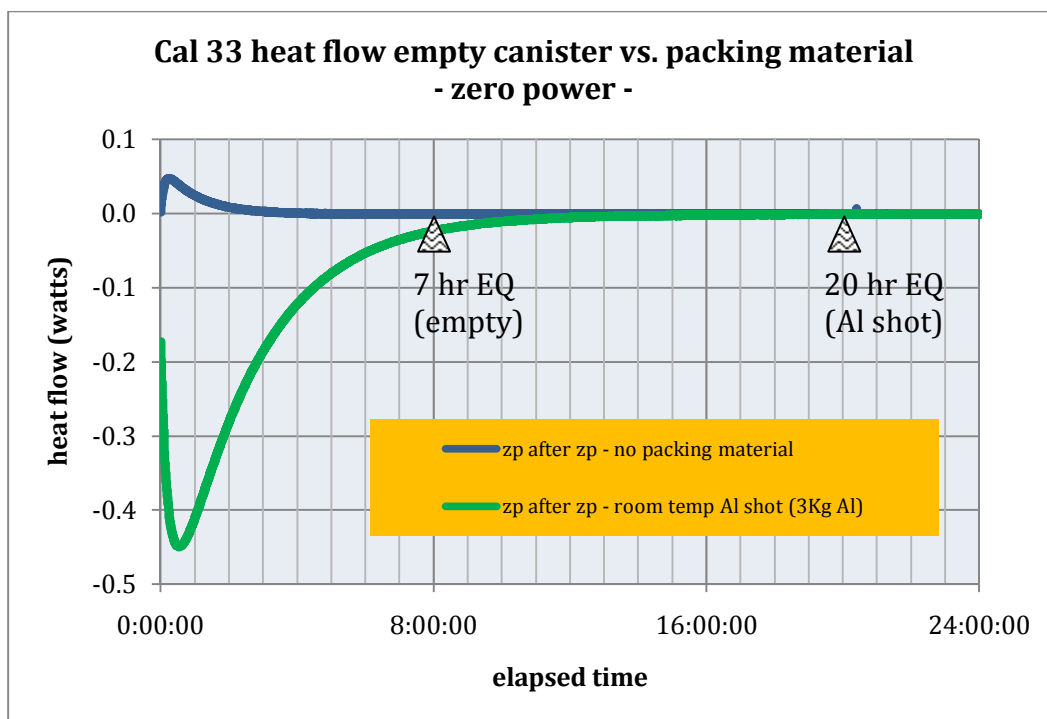


Figure 7. EQ time reduced 13 hours with no heat for empty canister vs. packing

### Considerations for Geometry and Contact

The impact of reducing unnecessary packing material from the sample canister made it clear that geometry and contact were far less significant factors than resistance for reducing thermal impedance. As far as geometry goes, our previous practice of centering samples by surrounding them with packing material caused us to increase mass and extend measurement times without benefit. The intent was to reduce unfavorable thermal gradients by making the distance to the chamber wall ( $R_{Th}$ ) as uniform as possible through the packing mass. However, the limited value of such an effort is borne out by the fact that the standards runs performed without any packing whatsoever were well within the limits of measurement control. For confirmation we compared the effect of centering a standard in the middle of a sample can filled with material against a run with the same standard and material where the can was located at the bottom of the sample canister. Both runs were preceded by zero-powers. The standard run that was centered came up to equilibrium in virtually the same time as when it was off-center and touching the inside bottom of the canister. The results for both runs were within control limits.

Although contact does not appear to be the major factor in measurement times, it is still an issue because of the extensive labeling that covers the outer surface of the sample containers. Some do not have much exposed metal surface at all. The aluminum balls and shot are likely to contact at least some of the metal surface directly and form a conduction path even for containers that are nearly covered with labels. The trade off is that using the shot and balls adds unwanted mass. We

needed a way to obtain good contact while minimizing mass (e.g. resistance) and came up with a concept for a light weight bracket designed to force contact of the sample container with the inner walls of the loading canister.

### **A Spider for PIhGs**

We could not perform runs with items containing SNM without some type of packing to secure them inside of the canister and we felt that there were still time gains to be made by attending to thermal contact between the items and the canister walls. We wanted minimal weight with good conductivity. We had already rejected the idea of using metal foams and honeycomb materials on the grounds that they would be troublesome to implement and that they could damage the various requisite seals and labels that adorn the sample cans. The solution needed to be simple to use without any special training, and it needed to be allowable within the existing controls for safety and security.

Our answer was to make a one size-fits-all cap for the sample containers to position them and ensure thermal contact with the canister. The concept was originally shaped like a spider used to center a part in a machine lathe. Although the design of the spider evolved to look differently, the name stuck, (Figure 8). We soon learned that it needed spring-assist to keep it in place during loading.

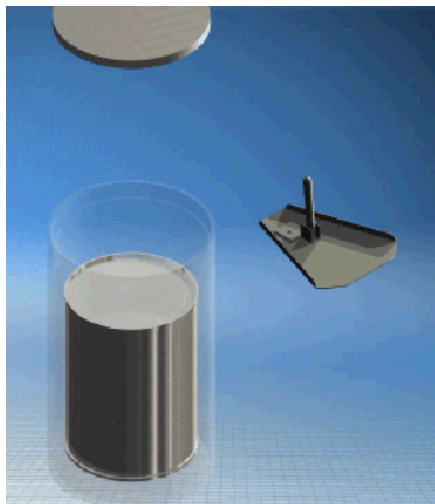


Figure 8. Spider, sample container, and canister



Spider in position

In testing, the PIhGs spider was demonstrated to reduce the run time of the 0.08 W standard in Cal 33 from 20 hours down to 7. For the 5.5 W standard, the time was reduced from 23 hours to 15. Both of these EQ times were the same as for an empty canister. The concept worked, but no additional time gains had been made.

## The PlhGs Trough

The figure below, (Figure 9), shows the masses of the packing materials that fit into the LLNL calorimeter loading canisters. The spiders are light enough that they are insignificant in terms of mass. The aluminum shot contributes a disproportionate mass given it is only used in the smallest canisters (cal's 32 and 33), and the foil balls add up to a surprisingly large amount in the large volume canisters despite their low average density of 0.13 g/cc; 500 to 3500 g (cal's 140a&b,18,29, and 30).

Fortunately, there is a simple, no cost, highly effective solution to reducing thermal impedance for all our configurations. Thermal contact with the canister walls is assured as well and the sample is supported safely. All that is required is to position the sample container so that it is touching the bottom of the canister and wall. Then, add just enough packing to hold it in place. We call this a trough. It allows for optimized thermal impedance in the simplest of ways and our testing shows that standards are measured within our limits of error.

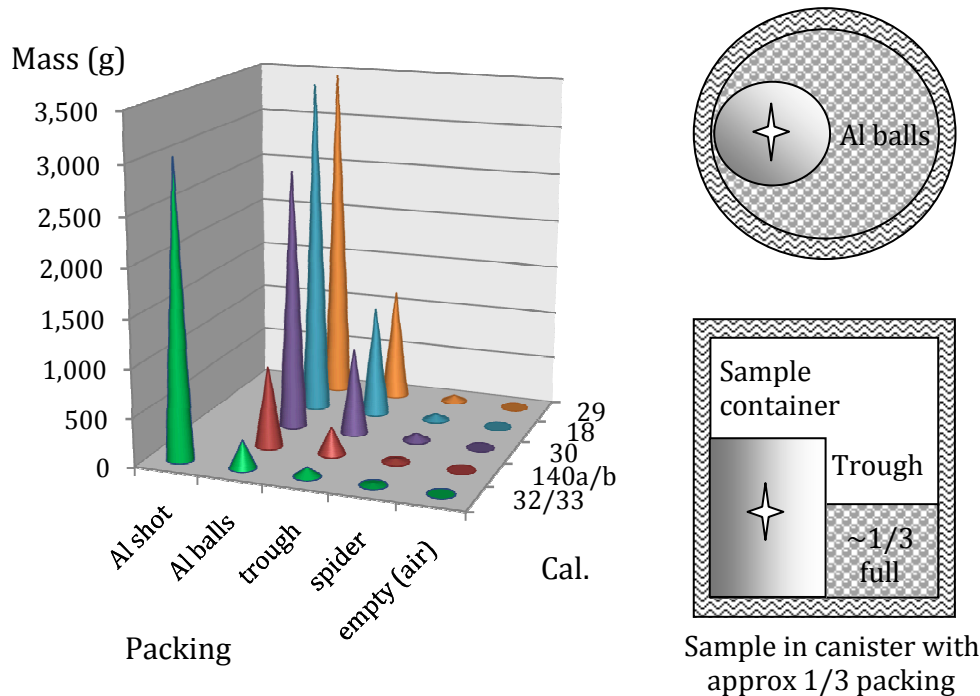


Figure 9. PlhGs troughs and spiders reduce packing mass significantly

Using the trough method reduces the mass of packing material to roughly 1/3<sup>rd</sup>, on average. The trough method is very workable in practice and operators find it the easiest method of packing. For small sample canisters it is possible to surpass performance of the spider because the total mass is low and the likelihood of establishing contact with more metal is increased, (Figure 10). For the large volume canisters, however, the spider mass can be much less than that for a trough. Further, the seals and labels that can get in the way on the small sample containers occupy less of the total surface area on the larger items.

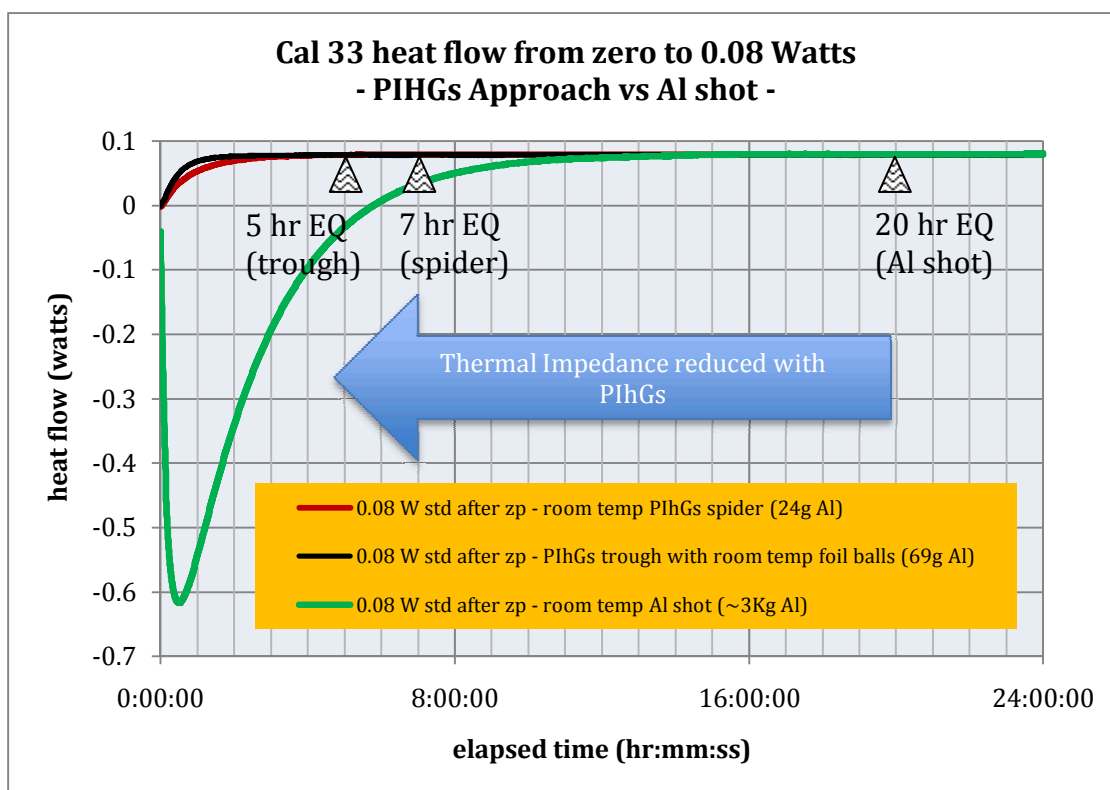


Figure 10. PIhGs trough surpasses spider performance vs. Al shot in small canisters

### End-Point Prediction

One of our hopes for reducing thermal impedance was that it would hasten the time to where a reasonably accurate prediction could be made for the heat-flow at equilibrium. “End-point prediction” involves real-time curve fitting of  $\Delta T$  readings. When the fit is sufficient, then the function is deemed to be predictable and the value at equilibrium is computed on faith. One problem with this approach is that the thermal gradients in system elements other than the known resistance ( $R_{Th}$ ), need to come to equilibrium as well before an accurate fit can be made. The various gradients mature at different times due to their differences in thermal resistance, their order in the chain of elements between the sample and the known resistance, and by the magnitude of the heat disturbance caused during sample loading. Reducing thermal impedance may or may not aid the prediction process any more than to shorten the overall time required. Instead, it may be more important to match the various resistances so that differences are minimized and the progression curve is smoother and easier to fit.

LLNL is not approved to use prediction schemes for inventory or standards measurements with any of its existing calorimeters nor does its existing equipment afford this capability. However, we have a new capability to investigate the effect of PIhGs sample packing principles in end-point prediction using a Setaram™ 3013

calorimeter that was procured in 2010. This new system has a feature that allows for end-point prediction but unfortunately, end-point experiments are postponed until the myriad of qualification runs for performing MC&A measurements in the standard mode are complete.

### **PIhGing Out**

Our efforts to reduce thermal impedance in the sample canisters lead to solutions that were useful in every one of our calorimeter systems. Measurement throughput was increased 25% overall. This gave us the extra we needed to meet all of the de-inventory milestones while working off a huge measurement backlog. As a result, the schedule remained on track and the NNSAs enhanced performance objectives were met. We were able to reduce the number of weekends that the Pu Facility had to be staffed which saved LLNL money and allowed workers to be home with their families on Saturday mornings again. In the final analysis, you could say that PIhGs saved enough bakin' to make the difference between success and failure; at least that's what the PIhGs spider did.

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This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.